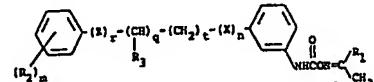


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 (71) Applicants
 Sumitomo Chemical
 Company Limited,
 No. 15 Kitahama
 5-chome, Higashi-ku,
 Osaka-shi, Osaka-fu,
 Japan
 (72) Inventors
 Yoshiaki Kosuge
 Ryo Yoshida
 Selzo Sumida
 Horofumi Oshita
 Soji Otsuki
 Katsuzo Kamoshita
 (74) Agents
 Boul, Wade & Tennant

(54) Oximecarbamate derivatives and their use as herbicides

(57) Oximecarbamate derivatives of the formula:



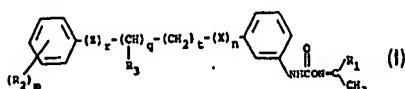
wherein R₁ is methyl or methoxy, R₂ is lower alkyl, lower alkoxy, methylenedioxy, methylthio, halogen or trifluoromethyl, R₃ is hydrogen, methyl or ethyl, X and Z are each oxygen or sulfur, m is an integer from 0 to 5 when R₂ is fluorine or an integer from 0 to 3 when R₂ is other than fluorine when m is 2 or 3, each R₂ is the same as or different from any other R², n and r are each 0 or 1 but are not both zero simultaneously, q is 0 or 1 and t is an integer from 0 to 4, which are useful as selective herbicides.

SPECIFICATION

Oximecarbamate derivatives and their use as herbicides

5 The present invention relates to oximecarbamate derivatives, and their production and use. More particularly, it relates to oximecarbamate derivatives of the formula:

10



wherein R₁ is methyl or methoxy, R₂ is lower alkyl, 15 lower alkoxy, methylenedioxy, methylthio, halogen or trifluoromethyl, R₃ is hydrogen, methyl or ethyl, X and Z are each oxygen or sulfur, m is an integer from 0 to 5 when R₂ is fluorine or an integer from 0 to 3 when R₂ is other than fluorine, and when m is 2 or 3 20 each R₂ is the same as or different from any other R², n and r are each 0 or 1 but are not both zero simultaneously, q is 0 or 1 and t is an integer from 0 to 4, it being understood that the word "integer" as used herein includes zero.

25 As used above, the term "lower" usually indicates a group having not more than 5 carbon atoms. Examples of lower alkyl are methyl, ethyl, n-propyl, isopropyl and t-butyl. Examples of lower alkoxy are methoxy, ethoxy, n-propoxy, isopropoxy and 30 t-butoxy. Examples of halogen are fluorine, chlorine and bromine.

The compounds of the invention are useful as herbicides for controlling and exterminating weeds without any unfavorable influence on the growth of 35 crop plants.

It is of course desirable that herbicides should have high safety to crop plants. However, herbicides for post-emergence application contact not only the foliage of weeds but also the foliage of crop plants 40 so that the extermination of only the weeds is very difficult. In fact few herbicides of this type are commercially available.

It has now been found that the oximecarbamate derivatives (I) of the present invention can control 45 and exterminate weeds efficiently without causing any chemical injury to crop plants such as soybean, cotton, sugar beet and wheat by post-emergence application. For instance, the compounds (I) can exterminate redroot pigweed (*Amaranthus retroflexus*), sunflower (*Helianthus annuus*), cocklebur (*Xanthium pennsylvanicum*), annual morningglory (*Ipomoea purpurea*), crabgrass (*Digitaria adscendens*), barnyard grass (*Echinochloa crus-galli*), etc. in the cotton or soybean field, and common lambs-quarter (*Chenopodium album*), radish, black nightshade (*Solanum nigrum*), green foxtail (*Setaria viridis*), etc. in the sugar beet or wheat field with high safety to the crop plants, when applied after emergence of the weeds. Thus, they are useful as 55 selective herbicides for post-emergence treatment in the culture of cotton, soybean, sugar beet or wheat.

Furthermore, the compounds (I) show a herbicidal activity by soil treatment of paddy fields. For instance, their application to rice paddy fields can 60 exterminate barnyard grass (*Echinochloa crus-galli*),

pickerel weed (*Monochoria vaginalis*), false pimpernel (*Lindernia pyxidaria*), toothcup (*Rotala indica*), nutssedge sp. (*Cyperus difformis*), etc. without causing any chemical injury to rice plants.

70 While the oximecarbamate derivatives (I) are novel, there is known O-(N-phenylcarbamoyl) acetoxime (W. German patent 1,024,746; Control (a)). However, no herbicidal activity of this control compound (a) on foliage treatment has been

75 described, and in fact, its herbicidal activity on foliage treatment is much inferior to that of the compounds (I).

The compounds are characteristic in having a substituent such as substituted phenoxy, substituted

80 phenylthio, substituted phenylalkyloxy, substituted phenylalkylthio, substituted phenoxyalkyl, substituted phenylthioalkyl, substituted phenoxyalkyloxy or phenylthioalkyloxy at the m-position of the phenyl group in the O-(N-phenylcarbamoyl)oxime derivative.

85 As will be shown hereinafter, the herbicidal activity of O-[N-{4-(2-fluorophenoxy-methyl)phenyl} carbamoyl] acetoxime (Control (b)) or of O-[N-{4-(3-(4-chlorophenyl)propoxy)phenyl} carbamoyl] acetoxime (Control (c)) is

90 much inferior to the herbicidal activity of the compounds (I).

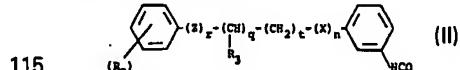
In general, the compounds (I) exhibit high selectivity to soybean, cotton, sugar beet and wheat. The selectivity is closely associated with their structure.

95 For instance, O-[N-{3-(3-chlorophenoxy-methyl)phenyl} carbamoyl] acetoxime (Compound No. 21) shows selectivity to cotton and sugar beet. When the chlorine atom at the m-position is changed to a trifluoromethyl group, the resulting

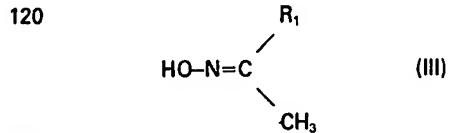
100 compound (Compound No. 32) shows selectivity to soybean and wheat losing selectivity to sugar beet and cotton. Further, when the methyl group in the dimethylacetoxime group of Compound No. 32 is replaced by a methoxy group, the resultant com-

105 pound (Compound No. 33) shows selectivity to sugar beet and wheat losing selectivity to soybean. Furthermore, O-[N-{3-(3-chlorophenethoxy-phenyl} carbamoyl] methoxyethanaloxime (Compound No. 103) shows selectivity only to wheat.

110 The compounds (I) are novel and can be produced, for instance, by reacting a phenylisocyanate derivative of the formula:



115 wherein R₂, R₃, X, Z, m, n, r, q and t are each as defined above, with an oxime derivative of the formula:



120 wherein R₁ is as defined above.

The reaction may be carried out in an inert organic solvent (e.g. benzene, toluene, xylene, diethyl ether, tetrahydrofuran, dioxane, N,N-dimethylformamide,

130 chloroform, carbon tetrachloride). The presence of a

tertiary amine (e.g. pyridine, triethylamine, diethylalanine) in the reaction system is advantageous for effecting the reaction efficiently. Usually, the oxime derivative (III) is employed in an equimolar to excess 5 molar amounts, preferably in an equimolar to 1.5 fold molar excess, to the phenylisocyanate derivative (II).

The reaction is usually effected at a temperature from 0 to 100°C and sometimes under cooling or heating, and it is normally completed within about 10 hours. The reaction product can be recovered from the reaction mixture by a conventional separation procedure such as filtration or distillation. If necessary, the recovered product may be further purified, for instance, by recrystallization or column chromatography.

The phenylisocyanate derivative (II) and the oxime derivative (III) are known.

Practical and presently preferred embodiments of
20 the preparation of the compounds (I) are illustra-
tively shown in the following examples.

Example 1

To a solution of acetoxime (8.1 g) in benzene (100 ml), there was dropwise added a solution of m - (3, 4 dichlorophenoxyethyl)phenylisocyanate (29.4 g) in benzene (50 ml) at 10 to 20°C. The mixture was stirred at the same temperature for 3 hours, and thereafter the solvent was removed. The residue was recrystallized from a mixture of benzene and tetrahydrofuran (5:1 by volume) to obtain 25.3 g of white needles (Compound No. 36). M.P., 79-81°C.

Elementary analysis: Calcd. for $C_{17}H_{18}Cl_2N_2O_4$: C, 55.59%; H, 4.40%; N, 7.63%; Cl, 19.30%. Found: C, 55.56%; H, 4.25%; N, 7.70%; Cl, 19.44%.

35 Example 2

To a solution of methoxyethanaloxime (8.9 g) in toluene (150 ml), there was added triethylamine (0.2 g). To this solution, there was dropwise added a solution of m - (4 - tert - butylphenoxy methyl) phenylisocyanate (28.1 g) in toluene (60 ml) at 50-60°C. The mixture was stirred at the same temperature for 2 hours, and thereafter the solvent was removed. The oily substance obtained was purified by column chromatography (silica gel, 70-230 mesh) using a mixture of benzene and tetrahydrofuran (8:1 by volume) to obtain 16.7 g of an oily substance (Compound No. 27).

50 Elementary analysis: Calcd. for $C_{21}H_{26}N_2O_4$: C, 68.07%; H, 7.09%; N, 7.56%. Found: C, 68.13%; H, 7.12%; N, 7.44%.

Some specific examples of the compound (I), which can be prepared in the same manner as above, are shown in Table 1 below but the compounds of the invention are not limitative to these examples.

Table 1

Compound No.	Chemical structure
1	
2	

Melting
point or
refrac-
tive index

M.P., 119-
120°C

CH₃
CH₃

M.P., 77-
79°C

CH₃
CH₃

Compound No.	Chemical structure	Melting point or refractive index M.P., 71-75°C
3		
4		n_D^{23} 1.5445
5		n_D^{23} 1.5312
6		$n_D^{25.5}$ 1.5910
7		$n_D^{25.5}$ 1.5882
8		M.P., 58-60°C
9		M.P., 119-121°C
10		M.P., 107-108°C
11		M.P., 59-60°C
12		n_D^{23} 1.5735
13		M.P., 95.5-96.5
14		M.P., 80-82°C
15		n_D^{24} 1.5671
16		M.P., 47.5-49.5°C
17		M.P., 89-91°C
18		M.P., 97-98°C
19		M.P., 10-13°C
20		n_D^{26} 1.569
21		M.P., 59-61°C

Compound No.	Chemical structure	Melting point or refractive index	Compound No.	Chemical structure	Melting point or refractive index
22		M.P., 76-77°C	40		M.P., 89-90°C
23		M.P., 76-77°C	41		M.P., 83-85°C
24		n_D^{24} 1.5821	42		M.P., 84-86°C
25		$n_D^{22.5}$ 1.5478	43		M.P., 45-50°C
26		M.P., 98-100°C	44		$n_D^{25.5}$ 1.5719
27		n_D^{25} 1.5590	45		n_D^{25} 1.5891
28		n_D^{24} 1.5771	46		M.P., 83-84°C
29		n_D^{21} 1.5800	47		M.P., 115-117°C
30		n_D^{23} 1.5645	48		M.P., 110-112°C
31		M.P., 81-82°C	49		$n_D^{25.5}$ 1.5305
32		n_D^{27} 1.5496	50		M.P., 116-117°C
33		n_D^{24} 1.5852	51		M.P., 77-78°C
34		n_D^{24} 1.5793	52		M.P., 79.5-81°C
35		$n_D^{25.5}$ 1.5341	53		M.P., 128-129°C
36		M.P., 79-81°C	54		M.P., 120-122°C
37		M.P., 36-37°C	55		M.P., 108-110°C
38		n_D^{25} 1.5778	56		M.P., 126-127°C
39		$n_D^{27.5}$ 1.5750	57		$n_D^{25.5}$ 1.5919
			58		n_D^{24} 1.5975

In the practical usage of the compounds (I), they may be applied neat or in the form of any composition such as wettable powders, emulsifiable concentrates, granules, fine granules or dusts.

5 In producing such compositions form, a solid or liquid carrier may be used. As examples of solid carriers, there may be given mineral powders (e.g. kaolin, bentonite, clay, montmorillonite, talc, diatomaceous earth, mica, vermiculite, gypsum, cal-
10 cium carbonate, apatite), vegetable powders (e.g. soybean powder, flour, wooden powder, tobacco powder, starch, crystalline cellulose), high molecular weight compounds (e.g. petroleum resin, polyvinyl chloride, dammar gum, ketone resin), alumina, wax
15 and the like.

Examples of liquid carriers are alcohols (e.g. methanol, ethanol, ethylene glycol, benzyl alcohol), aromatic hydrocarbons (e.g. toluene, benzene,

xylene, methylnaphthalene), halogenated hydrocarbons (e.g. chloroform, carbon tetrachloride, monochlorobenzene), ethers (e.g. dioxane, tetrahydrofuran), ketones (e.g. acetone, methyl ethyl ketone, cyclohexanone), esters (e.g. ethyl acetate, butyl acetate, ethylene glycol acetate), acid amides (e.g. dimethylformamide), nitriles (e.g. acetonitrile), ether alcohols (e.g. ethylene glycol ethyl ether), water and the like.

A surface active agent may be used for emulsification dispersion or spreading and may be any of the

30 non-ionic, anionic, cationic and amphoteric type of agents. Examples of the surface active agent include polyoxyethylene alkyl ethers, polyoxyethylene alkylaryl ethers, polyoxyethylene fatty acid esters, sorbitan fatty acid esters, polyoxyethylene sorbitan

35 fatty acid esters, oxyethylene polymers, oxypolypropylene polymers, polyoxyethylene alkyl phosphates, fatty acid salts, alkyl sulfates, alkyl sulfonates, alkylaryl sulfonates, alkyl phosphates, quaternary ammonium salts and the like. But, the surface

40 active agent is not of course limited to these compounds. And, if necessary, gelatin, casein, sodium alginate, starch, agar, polyvinyl alcohol or the like may be used as an auxiliary agent.

In the preparation of a herbicidal composition, the 45 content of the compound (I) may be from 1 to 95% by weight, preferably from 1 to 80% by weight.

Practical embodiments of the herbicidal composition according to the invention are illustratively shown in the following examples wherein parts and % are by weight.

Preparation Example 1

Eighty parts of Compound No. 36, 5 parts of polyoxyethylene alkylaryl ether and 15 parts of synthetic silicon oxide hydrate are well mixed while being powdered to obtain a wettable powder.

Preparation Example 2

Thirty parts of Compound No. 21, 7 parts or polyoxyethylene alkylaryl ether, 3 parts of alkylaryl sulfonate and 60 parts of xylene are well mixed to obtain an emulsifiable concentrate.

Preparation Example 3

One part of Compound No. 69, 1 part of white carbon, 5 parts of ligninsulfonate and 93 parts of clay are well mixed while being powdered. The mixture is then well kneaded with water, granulated and dried to obtain a granule.

Preparation Example 4

Fourty parts of bentonite, 5 parts of ligninsulfonate and 55 parts of clay are well mixed while being powdered. The mixture is then well kneaded with water, granulated and dried to obtain a granule containing no active ingredient. The granule is then impregnated with 5 parts of Compound No. 32 to obtain a granule.

75 Preparation Example 5

Three parts of Compound No. 27, 0.5 part of isopropyl phosphate, 66.5 parts of clay and 30 parts of talc are well mixed while being powdered to obtain a dust.

may be given phenoxy series herbicides such as 2, 4 - dichlorophenoxyacetic acid, 2 - methyl - 4 - chlorophenoxyacetic acid and 2, 4 - dichlorophenoxybutyric acid (including esters and salts thereof);

5 diphenyl ether series herbicides such as 2, 4 - dichlorophenyl - 4' - nitrophenyl ether, 2, 4, 6 - trichlorophenyl - 4' - nitrophenyl ether, 2, 4 - dichloro - 3' - methoxy - 4' - nitrophenyl ether, 2, 4 - dichlorophenyl - 3' - methoxycarbonyl - 4' - nitrophenyl ether and 2 -

10 chloro - 4 - trifluoromethylphenyl - 3' - hydroxycarbonyl - 4' - nitrophenyl ether; triazine series herbicides such as 2 - chloro - 4, 6 - bisethylamino - 1, 3, 5 - triazine, 2 - chloro - 4 - ethylamino - 6 - isopropylamino - 1, 3, 5 - triazine, 2 - methylthio - 4, 6 -

15 bisethylamino - 1, 3, 5 - triazine, 2 - methylthio - 4, 6 - bisisopropylamino - 1, 3, 5 - triazine, 4 - amino - 3 - methyl - 6 - phenyl - 1, 2, 4 - triazine - 5 (4H) - one and 4 - amino - 6 - tert - butyl - 3 - methylthio - 1, 2, 4 - triazine - 5 (4H) - one; urea series herbicides such as

20 3 - (3, 4 - dichlorophenyl) - 1, 1 - dimethylurea, 3 - (3, 4 - dichlorophenyl) - 1 - methoxy - 1 - methylurea, 3 - (3 - chloro - 4 - difluorochloromethylthiophenyl) - 1, 1 - dimethylurea, 3 - [4 - (4 - chlorophenoxy)phenyl] - 1, 1 - dimethylurea and 3 - (α, α, α - trifluoro - m - tolyl) -

25 1, 1 - dimethylurea; carbamate series herbicides such as isopropyl - N - (3 - chlorophenyl)carbamate, methyl - N - (3, 4 - dichlorophenyl) carbamate and 4 - chloro - 2 - butynyl - m - chlorocarbanilate; thiocarbamate series herbicides such as S - (4 -

30 chlorobenzyl) - N, N - diethylthiocarbamate, S - ethyl - N, N - hexamethylenethiocarbamate and S - ethyl dipropylthiocarbamate; acid anilide series herbicides such as 3, 4 - dichloropropionanilide, N - methoxymethyl - 2, 6 - diethyl - 2 - chloroacetanilide

35 and 2 - chloro - 2', 6' - diethyl - N - (butoxymethyl)acetanilide; uracil series herbicides such as 5 - bromo - 3 - sec - butyl - 6 - methyluracil and 3 - cyclohexyl - 5, 6 - trimethylureauracil; pyridinium salt series herbicides such as 1, 1' -

40 dimethyl - 4, 4' - bispyridinium dichloride; phosphorus series herbicides such as N - (phosphonomethyl)glycine, O - methyl - O - (2 - nitro - 5 - methylphenyl) - N - sec - butylphosphoroamidothioate and O - ethyl - O - (2 -

45 nitro - 4 - methylphenyl) - N - isopropyl - phosphoroamidothioate; toluidine series herbicides such as α, α, α - trifluoro - 2, 6 - dinitro - N, N - dipropyl - p - toluidine, N - (cyclopropylmethyl) - α, α, α - trifluoro - 2, 6 - dinitro - N - propyl - p - toluidine; N

50 - sec - butyl - 4 - tert - butyl - 2, 6 - dinitroaniline; 3, 5 - dinitro - 4 - N, N - dipropylaminosulfanylamide; 5 - tert - butyl - 3 - (2, 4 - dichloro - 5 - isopropoxyphenyl) - 1, 3, 4 - oxadiazolin - 2 - one; 3 - isopropyl - 1H - 2, 1, 3 - benzothiadiazine(4) - 3H - one - 2, 2 - dioxide

55 (including salts thereof); α - (β - naphthoxy)propionanilide; 2 - (α - naphthoxy) - N, N - diethylpropionamide; 3 - amino - 2, 5 - dichlorobenzoic acid; 2 - sec - butyl - 4, 6 - dinitrophenol; N - 1 - naphthylphthalamic acid; 5 -

60 amino - 4 - chloro - 2 - phenyl - 3 (2H) - pyridazine and the like. But, the herbicides are not of course limited to these examples.

The herbicides of the invention may be applied together with fungicides, pyrethroid series insecticides, other insecticides, plant growth regulators,

fertilizers, etc.

The dosage rate of the compounds (I) depends upon their kinds, the sorts of cultivated plants, the method of application, etc. Generally, however, the dosage rate is from 2 to 200 grams, preferably from 5 to 50 grams, of the active ingredient per are.

The application of the compounds (I) as herbicides will be illustrated in the following Examples wherein the phytotoxicity to cultivated plants and the herbicidal activity on weeds were evaluated as follows: the aerial parts of the test plants were cut off and weighed (fresh weight); the percentage of the fresh weight of the treated plant to that of the untreated plant was calculated with the latter fresh weight taken as 100; and the crop damage and the herbicidal activity were evaluated by the standard given in the table below. The rating values of phytotoxicity, 0 and 1, and those of herbicidal effect, 5 and 4, are generally regarded as satisfactory to protect cultivated plants and to control weeds, respectively. The rating values in the paddy rice test alone were calculated from the dry weight of plant.

Rating value	Fresh weight (percentage to untreated plot)	
	Cultivated plant	Weed
5	0 - 39	0
4	40 - 59	1 - 10
3	60 - 79	11 - 20
2	80 - 89	21 - 40
1	90 - 99	41 - 60
0	100	61 - 100

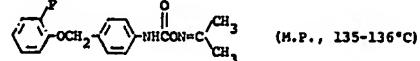
The following control compounds were used in the Examples.

100 Control (a)



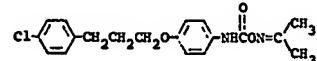
(M.P., 143-145°C)

105 Control (b)



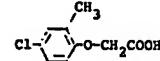
(M.P., 135-136°C)

110 Control (c)



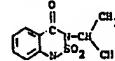
(M.P., 120-121°C)

115 HCP

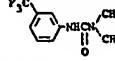


HCP

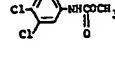
120 Pentaxon



125 Pluonaturon



130 SWEP



Example 1 (Paddy race test)

Wagner's pots (1/5000 are) were each filled with 1.5 kg of paddy field soil containing the seeds of weeds and kept under flooded conditions. The seedlings of rice plant at a 3-leaf stage were transplanted thereto, and after the seeds of barnyard grass were sowed therein, the seedlings were grown for 15 days in a green-house. Thereafter, the required amount of the wettable powder of each test compound was 10 diluted with water and applied to the soil under flooded conditions. Twenty-five days after the application, the evaluation of herbicidal activity and phytotoxicity was made on the rice plants and barnyard grass as well as broad-leaved weeds (e.g. 15 kерel weed (*Monochoria vaginalis*), false pimpernel (*Lindernia pyxidaria*), toothcup (*Rotala indica*) and nutsedge sp. (*Cyperus difformis*)). The results are shown in Table 2.

Table 2

Compound No.	Dosage (weight of active ingredient, g/arc)	Evaluation of crop damage and herbicidal activity			
		Rice plant	Barn-yard grass	Broad-leaved wood	Nutsedge sp.
1	20	0	4	4	5
2	20	0	4	5	5
3	20	0	5	5	5
	10	0	4	5	4
4	20	0	4	5	5
5	20	0	4	4	4
6	20	0	-	5	4
7	20	0	4	5	5
8	20	0	-	5	5
9	20	0	-	5	4
10	20	0	4	5	5
11	20	0	-	5	5
12	20	0	4	5	5
13	20	0	-	5	5
14	20	0	5	5	5
15	20	0	-	5	5
16	20	0	4	5	5
17	20	0	5	5	4
18	20	0	5	5	5
	10	0	4	5	5
19	20	0	5	5	5
	10	0	5	5	5
20	20	0	5	5	5
	10	0	5	5	5
21	20	0	5	5	5
	10	0	4	5	5
22	20	0	5	5	5
	10	0	4	5	5
23	20	0	-	5	5
24	20	0	4	4	5
25	20	1	5	5	5
26	20	0	5	5	5
	10	0	4	5	5
27	20	0	5	5	5
28	20	0	4	5	4
29	20	0	4	5	5
30	20	0	-	5	5
31	20	0	-	4	4
32	20	0	5	5	5
	10	0	5	5	5
33	20	0	4	5	5
35	20	0	5	5	5
	10	0	4	5	5
36	20	0	5	5	5
	10	0	4	5	5
37	20	0	5	5	5
	10	0	5	5	5
38	20	0	5	5	5
	10	0	4	5	5
39	20	0	4	5	4
40	20	0	4	5	5
	10	0	-	5	4

(Continued)

Compound No.	Dosage (weight of active ingredient, g/arc)	Evaluation of crop damage and herbicidal activity			
		Rice plant	Barn-yard grass	Broad-leaved wood	Nutsedge sp.
41	20	0	5	5	5
	10	0	4	5	5
42	20	0	-	5	4
43	20	0	-	4	4
44	20	0	5	5	5
45	20	0	5	5	5
	10	0	4	5	5
46	20	0	4	5	5
47	20	0	-	5	5
48	20	0	4	5	4
49	20	0	5	5	5
50	20	0	5	5	5
51	20	0	5	5	5
	10	0	4	5	5
52	20	0	5	5	5
53	20	0	4	5	5
54	20	0	4	5	5
55	20	0	5	5	5
	10	0	4	5	5
56	20	0	5	5	5
	10	0	4	5	5
57	20	0	5	5	5
	10	0	5	5	5
58	20	0	-	5	4
59	20	0	5	5	5
	10	0	4	5	5
60	20	0	5	5	5
61	20	0	-	5	5
62	20	0	-	5	5
63	20	0	4	4	4
64	20	0	4	5	5
65	20	0	4	5	5
66	20	0	5	5	5
67	20	0	-	4	5
68	20	0	-	5	5
69	20	0	4	5	5
70	20	0	4	5	4
71	20	0	4	5	5
72	20	0	-	5	4
73	20	0	-	5	5
74	20	0	-	4	4
75	20	0	4	5	4
76	20	0	-	4	4
77	20	0	4	5	5
78	20	0	-	5	4
79	20	0	-	4	4
80	20	0	-	5	5
81	20	0	4	5	5
82	20	0	4	5	5
83	20	0	4	5	5
84	20	0	5	5	5
	10	0	4	5	5
85	20	0	1	5	5
	10	0	0	4	4
86	20	0	0	4	4
87	20	0	0	4	5
88	20	0	0	-	4
89	20	0	0	-	5
90	20	0	0	-	5
91	20	0	0	-	5
92	20	0	0	5	5
	10	0	0	4	5
93	20	0	0	-	5
94	20	0	0	-	5
95	20	0	0	4	5
96	20	0	0	4	5
97	20	0	0	-	5
98	20	0	0	4	5
	10	0	0	4	5
99	20	0	0	4	5
100	20	0	0	4	5
101	20	0	0	4	5
	10	0	0	4	5
102	20	0	0	4	5

(Continued)

Compound No.	Dosage (weight of active ingredient, g/are)	Evaluation of crop damage and herbicidal activity			
		Rice plant	Barnyard grass	Broad-leaved weed	Nutsedge sp.
103	20	0	5	5	5
	10	0	4	5	5
104	20	0	4	5	5
105	20	0	-	5	5
106	20	0	-	5	4
107	20	0	-	5	4
108	20	0	4	5	5
109	20	0	4	5	5
110	20	0	-	5	4
Control (a)	40	0	0	2	1
	20	0	0	1	0
Control (b)	40	1	0	3	2
	20	0	0	2	0
Control (c)	40	1	1	3	3
	20	1	0	1	2
MCP	20	3	4	5	5
	10	3	3	5	5

Example II (Post-emergence application test (weeds))

Plastic trays (35 cm x 25 cm x 10 cm (high)) were filled with upland field soil, and the seeds of redroot 5 pigweed, common lambsquarter, radish, sunflower, cocklebur, annual morningglory, black nightshade, large crabgrass, barnyard grass and green foxtail were separately sowed in the trays and grown for 3 weeks in a green-house. The required amount of the 10 test compound was sprayed to the foliage of the test plants over the top by means of a small hand sprayer. After the spraying, the test plants were grown for further 3 weeks in the green-house, and herbicidal activity was examined. The results are 15 shown in Table 3. In the above foliar application, the test compounds were each formulated into an emulsifiable concentrate, and the required amount of the emulsifiable concentrate was dispersed in water for application at a spray volume of 5 liters per are and 20 applied with the addition of a wetting agent. At the time of application, the weeds were in a 2- to 4-leaf stage and 2 to 12 cm in height although there was some difference depending upon the kind of weed.

Table 3

Compound No.	Dosage (weight of active ingredient, g/are)	Herbicidal activity									
		Redroot pigweed	Common lambs-quarter	Radish	Sunflower	Cocklebur	Annual morning-glory	Black nightshade	Large crabgrass	Barnyard grass	Green foxtail
3	20	5	5	5	5	5	5	5	4	4	4
4	20	5	5	5	5	5	4	5	4	-	4
6	20	5	5	5	5	5	4	5	-	-	4
7	20	5	5	5	5	5	5	5	-	4	4
8	20	5	5	5	5	5	5	5	5	-	4
	10	5	5	5	5	5	5	5	4	-	4
10	20	5	5	5	5	5	4	5	-	4	4
12	20	5	5	5	5	5	5	-	-	-	4
14	20	5	5	5	5	5	5	5	-	5	-
15	20	5	5	5	5	5	5	5	4	5	4
16	20	5	5	5	5	5	5	5	-	5	4
	10	5	5	5	5	5	5	5	-	5	4
17	20	5	5	5	5	5	5	5	5	4	5
18	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	-	4	5
19	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	4	4	5
20	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	5	4	4
21	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	-	5	4
22	20	5	5	5	5	5	5	5	5	4	5
23	20	5	5	5	5	5	5	4	5	4	4
26	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	4	4	4
27	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	-	4	4
29	20	5	5	5	5	5	5	5	4	4	4
32	20	5	5	5	5	5	5	5	5	4	4
	10	5	5	5	5	5	5	5	-	5	4
33	20	5	5	5	5	5	5	5	4	5	4
	10	5	5	5	5	5	5	5	-	5	4
35	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	-	4	5
36	20	5	5	5	5	5	5	5	5	4	5
	10	5	5	5	5	5	5	5	-	4	4
37	20	5	5	5	5	5	5	5	5	4	5
	10	5	5	5	5	5	5	5	-	4	5
38	20	5	5	5	5	5	5	5	4	4	4
	10	5	5	5	5	5	5	5	-	4	4
40	20	5	5	5	5	5	5	5	4	4	4
41	20	5	5	5	5	5	5	5	4	4	5
44	20	5	5	5	5	5	5	5	4	5	4
	10	5	5	5	5	5	5	5	-	4	4
45	20	5	5	5	5	5	5	5	4	4	4
	10	5	5	5	5	5	5	5	-	4	4
46	20	5	5	5	5	5	5	5	4	-	4
49	20	5	5	5	5	5	5	5	4	5	5
51	20	5	5	5	5	5	5	5	4	4	4
52	20	5	5	5	5	5	5	5	4	5	4
	10	5	5	5	5	5	5	5	-	4	4

(Continued)

Compound No.	Dosage (weight of active ingredient, g/are)	Herbicidal activity									
		Redroot pigweed	Common lambs-quarter	Radish	Sunflower	Cocklebur	Annual morning-glory	Black nightshade	Large crab-grass	Barnyard grass	Green foxtail
53	20	5	5	5	5	5	5	5	4	5	4
54	20	5	5	5	5	5	5	5	4	4	5
55	20	5	5	5	5	5	5	5	4	5	5
56	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	4	5	5
57	20	5	5	5	5	5	5	5	5	4	5
	10	5	5	5	5	5	5	5	4	4	4
58	20	5	5	5	5	5	4	5	4	—	4
59	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	4	4	5
60	20	5	5	5	5	5	5	5	5	4	5
	10	5	5	5	5	5	5	5	4	—	4
61	20	5	5	5	5	5	5	5	—	5	5
63	20	5	5	5	5	5	5	5	4	4	5
64	20	5	5	5	5	5	5	5	4	5	4
67	20	5	5	5	5	5	5	5	4	4	5
69	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	5	4	4
70	20	5	5	5	5	5	5	5	—	4	5
71	20	5	5	5	5	5	5	5	4	4	5
72	20	5	5	5	5	5	5	5	4	5	5
75	20	5	5	5	5	5	5	5	—	4	—
76	20	5	5	5	5	5	4	5	—	4	4
78	20	5	5	5	5	5	5	5	—	—	4
79	20	5	5	5	5	5	5	5	5	4	5
	10	5	5	5	5	5	5	5	5	4	4
80	20	5	5	5	5	5	5	5	—	4	4
81	20	5	5	5	5	5	5	5	4	4	5
83	20	5	5	5	5	5	5	5	4	4	4
84	20	5	5	5	5	5	5	5	—	4	5
85	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	4	—	5
87	20	5	5	5	5	5	4	5	—	4	4
91	20	5	5	5	5	5	5	5	4	—	5
92	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	4	3	3
94	20	5	5	5	5	5	5	5	—	—	4
95	20	5	5	5	5	5	5	5	4	—	4
96	30	5	5	5	5	5	5	5	4	—	—
97	20	5	5	5	5	5	5	5	4	5	5
98	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	5	4	5
99	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	4	4	5
100	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	—	4	4
101	20	5	5	5	5	5	5	5	4	4	4
	10	5	5	5	5	5	5	5	—	4	4
102	20	5	5	5	5	5	5	5	5	5	4
	10	5	5	5	5	5	5	5	—	4	4
103	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	4	4	5
104	20	5	5	5	5	5	5	5	4	4	—
105	20	5	5	5	5	5	5	5	—	4	4
108	20	5	5	5	5	5	5	5	5	5	5
	10	5	5	5	5	5	5	5	4	—	4
110	20	5	5	5	5	5	5	5	4	4	4
Control (a)	40	1	0	0	0	0	0	1	0	0	0
	20	0	0	0	0	0	0	0	0	0	0
Control (b)	40	0	0	1	0	0	0	0	0	0	0
	20	0	0	0	0	0	0	0	0	0	0
Control (c)	40	0	2	2	0	0	0	3	0	0	1
	20	0	0	0	0	0	0	1	0	0	0
Bentazon	20	3	5	5	5	5	4	5	1	2	0
	10	3	5	5	5	5	2	5	0	1	0
Fluometuron	20	5	5	5	5	5	5	5	4	5	5
	10	5	5	5	5	5	5	5	4	4	5
Sweg	20	5	5	5	5	5	3	5	2	4	4
	10	5	4	4	5	5	3	3	4	4	4

Example III (Post-emergence application test (cultivated plants))

Wagner's pots (1/5000 are) were each filled with upland field soil, and the seeds of soybean, cotton, sugar beet and wheat were sowed in the pots and grown for 3 weeks in a green-house. The required amount of the test compound was sprayed to the

foliage of the test plants over the top by means of a small hand sprayer. After the spraying, the test

plants were grown for further 3 weeks in the green-house, and phytotoxicity was examined. In the above foliar application, the test compounds were each formulated into an emulsifiable concentrate, and the required amount of the emulsifiable concentrate was

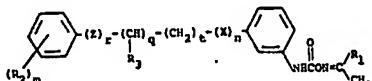
dispersed in water for application with the addition of wetting agent. At the time of application, soybean was in the second trifoliate stage, cotton in the 1-leaf stage, sugar beet in the 2-leaf stage and wheat in the 5 2-leaf stage. The results are shown in Table 4.

Table 4

Compound No.	Dose/g (weight of active ingredient, g/arc)	Phytotoxicity			
		Soybean	Cotton	Sugar beet	Wheat
3	20	1	-	-	1
7	20	1	-	-	-
8	20	-	-	-	10
	10	-	-	-	-
36	20	1	-	-	00
	10	1	-	-	00
37	20	-	-	1	10
	10	-	-	1	-
38	20	1	-	-	-
	10	1	-	-	-
41	20	1	-	-	0
44	20	1	-	-	10
	10	1	-	-	-
45	20	-	-	-	10
	10	-	-	-	-
46	20	1	-	1	0
49	20	0	1	-	-
52	20	1	1	-	-
	10	0	1	-	-
56	20	0	0	-	10
	10	0	0	-	-
61	20	1	-	-	00
	10	1	-	-	00
60	20	1	1	-	10
	10	0	1	-	-
69	20	1	-	-	-
	10	1	-	-	-
71	20	0	1	-	-
76	20	1	0	-	-
79	20	0	-	-	-
81	20	1	-	0	0
84	20	1	-	-	-
85	20	0	-	-	-
98	20	1	0	-	-
	10	0	0	-	-
99	20	-	-	-	00
	10	-	-	-	00
101	20	1	-	1	-
	10	0	-	1	-
102	20	1	-	1	-
	10	0	-	0	-
103	20	-	-	-	10
	10	-	-	-	-
Bentazon	20	0	-	5	-
	10	0	-	5	-
Fluomaturon	20	-	2	-	-
	10	-	1	-	-
Swop	20	-	-	5	3
	10	-	-	5	1

CLAIMS

1. A compound of the formula:



wherein R_1 is methyl or methoxy, R_2 is lower alkyl, lower alkoxy, methylenedioxy, methylthio, halogen or trifluoromethyl, R_3 is hydrogen, methyl or ethyl, X 15 and Z are each oxygen or sulfur, m is an integer from 0 to 5 when R_2 is fluorine or an integer from 0 to 3 when R_2 is other than fluorine when m is 2 or 3 each R_2 is the same as or different from any other R^2 , n and r are each 0 or 1 but are not both zero, simultaneously, q is 0 or 1 and t is an integer from 0 to 4.

2. A compound as claimed in claim 1, wherein R₁

is methoxy, R_2 is lower alkyl, lower alkoxy or halogen, X is oxygen, m is 0, 1 or 2, n is 1, r is 0, q is 1 and t is an integer from 1 to 4.

25 3. A compound as claimed in claim 1, wherein R_2 is lower alkyl, lower alkoxy, halogen or trifluoromethyl, R_3 is hydrogen or methyl, Z is oxygen, m is an integer from 0 to 3, n is 0, r is 1, q is 1 and t is 0.

30 4. Any one of compounds 1 to 110 as hereinbefore defined.

5. $O-[N-\{3-(3\text{-chlorophenoxy)methyl\} phenyl\} carbamoyl] acetoxime.$

6. $O-[N-\{3-(3\text{-trifluoromethylphenoxy\} methyl\} phenyl\} carbamoyl] acetoxime.$

35 7. $O-[N-\{3-(3\text{-trifluoromethylphenoxy\} methyl\} phenyl\} carbamoyl] methoxyethanoxime.$

8. $O-[N-\{3-(3,4\text{-dichlorophenoxy)methyl\} 40 phenyl\} carbamoyl] acetoxime.$

9. $O-[N-\{3-(3,5\text{-dichlorophenoxy)methyl\} phenyl\} carbamoyl] acetoxime.$

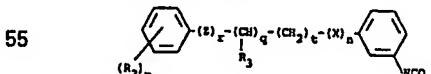
45 10. $O-[N-\{3-(3,4\text{-dichlorophenyl-3-propanoxy\} phenyl\} carbamoyl] methoxyethanoxime.$

11. $O-[N-\{3-(4\text{-methylphenethoxy\} phenyl\} carbamoyl] methoxyethanoxime.$

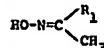
12. $O-[N-\{3-(3\text{-chlorophenethoxy\} phenyl\} carbamoyl] methoxyethanoxime.$

50 13. $O-[N-\{3-(4\text{-tert-butylphenyloxymethyl\} phenyl\} carbamoyl] methoxyethanoxime.$

14. A process for preparing a compound as claimed in claim 1, which comprises reacting a compound of the formula:



with a compound of the formula:



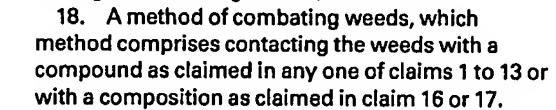
wherein $R_1, R_2, R_3, X, Z, m, n, q, r$ and t are each as defined in claim 1.

defined in claim 1.

15. A compound as claimed in claim 1 when prepared by a process as claimed in claim 14.

65 16. A herbicidal composition which comprises a compound as claimed in any one of claims 1 to 13 or claim 15, together with a carrier.

17. A composition as claimed in claim 16, in the form of a wettable powder, an emulsifiable concentrate, granules, fine granules, or a dust.



75 19. A method of selectively combating weeds in
the cultivation of soybean, cotton, sugar beet or
wheat, which comprises applying a compound as
claimed in any one of claims 1 to 13 or a composition
as claimed in claim 16 or 17, to the area wherein the
80 soybean, cotton, sugar beet or wheat is cultivated.

20. Use of the compound according to claim 1 as a herbicide.